



Identification of Sources and Mobilization of Trace Elements in Shallow Groundwater of the Upper Ganges River Basin

Md. Shajedul Islam^{1*}, M. G. Mostafa², Muhammad Touseef^{3,4}

¹Govt. HSS College, Magura 6700,
Ministry of Education, Bangladesh

²Institute of Environmental Science,
University of Rajshahi, Rajshahi 6205,
Bangladesh

³College of Civil Engineering and
Architecture, Guangxi University, Nanning
530004, China

⁴Guangxi Key Laboratory of Disaster
Prevention and Engineering Safety,
Nanning 530004, China

Correspondence: shajedulkst@gmail.com

Abstract

The concentration of trace metals in groundwater is still not within the usual standards established by the national and international monitoring authorities. This study aims to evaluate the sources and distribution of trace metals in the shallow aquifer water of the Ganges River basin in the Bangladesh area. A total of 40 groundwater samples were collected and investigated for 11 trace elements and some selected water parameters followed by sophisticated methods. The results presented that three metals, viz. Fe, Mn, and Pb crossed the limit of WHO standard in most of the samples (70-100%) representing severe human health hazards. Besides, the samples are slightly acidic to basic and highly mineralized by divalent cations and bicarbonate because total hardness was found very high. To assess factors affecting the trace metal loads in groundwater samples and determine their possible sources and mobility, multivariate statistical techniques, including Pearson's correlation coefficient, principal components analysis, and dendrogram cluster analysis are performed. Results revealed that sources of trace elements were identified to be geologic and little from human activities, i.e., application of agrochemicals, excess water mining, and industrial wastewater. It was recognized that the elevated concentration of trace elements in samples was caused by geogenic factors rather than manmade activities. Groundwater in this area poses a large threat due to high levels of trace metals, without any purification, it is essential to avoid this water for drinking. It advised that the groundwater quality should be constantly monitored for sustainable water quality management in the study zone.

Keywords: Aquifer basement, Geogenic process, Groundwater, Multivariate analysis, Rock weathering, Toxicity

1. Introduction

Recently, groundwater quality and quantity are drastically deteriorating in Bangladesh due to over-exploitation of water, excessive use of agrochemicals, urbanization, and vast industrialization [1]. Moreover, frequent changes in geogenic processes and some anthropogenic causes deteriorated massively the groundwater quality concerning trace and heavy metal pollution in some parts of the country during the

last 3-4 decades [2, 3]. Thus, the proper evaluation of sources and mobilization mechanisms of harmful trace metals such as Fe, Mn, Co, Cd, Pd, Cr, Hg, As, Zn, Cu, etc. (at a certain concentration) in aquifer basements is very important to ensure safe water for all-purpose uses. Groundwater contamination through trace elements is a global problem and a serious threat to public health, the natural ecosystem, food resources, and facilities [4]. Due to the consumption of groundwater, the

presence of toxic elements in human physiology can cause numerous problems. These elements in small amounts in the body may preserve cells. Though, the presence of more than the threshold limit of these elements may cause damage to the biotic organisms [5]. Trace metal analysis, distribution, and source identification in groundwater samples of the Bengal delta (Ganges basin) are the major aim of this study.

Trace metals have the property of ecosystem perseverance and bioaccumulation, and these metals pass in the aquatic system through several routes. These elements can be found on the earth's crust in their regular form. Some of them are very toxic and they not only harm the quality of the aquatic environment but also human well-being [6]. These metals are so hazardous that they cannot be degraded or decomposed in ecosystems and they have the ability to bioaccumulate [7]. Trace elements once get into the environment through the air, drinking water, or numerous chemicals and products that are manmade. The toxicity of these elements in the human body system decreases energy levels; disturbs brain functioning; disrupts the operation of numerous other organs such as the kidney, lungs, brain, and liver; and hampers blood composition.

Trace metals have two major sources in groundwater: (a) natural or geogenic and (b) anthropogenic or manmade sources. Natural sources include the dissolution of parent rocks at the aquifer level [8]. At first, these metals or their minerals accumulate in the soil layer, which is leached with rain or flood water and finally reaches the sub-surface water layer. The sources of trace metals in groundwater through human activities are well defined but the natural or geogenic activities are not easy to identify. These metals are found in high concentrations in the environment, they are formed by natural courses such as volcanic eruptions, dissolution of rocks/minerals in the water phase, and discharge into rivers, lakes, and oceans owing to the action of water [9]. Loads of trace metal in groundwater depend on the groundwater's local lithology or geology, hydrogeology, and geochemistry [10]. One of the chief geogenic sources of metal contamination is the dissolution of metal-laden rocks in groundwater. The

dissolution of sedimentary rocks such as calcite, dolomite, shale, or mixed rock makes the water polluted through the high level of toxic metals. When the interaction of water with rock elements occurs, it leads to the addition of these metals into the water phase; thus, pollution arises. Examples of such rocks are basalt, gabbro, granite, siderite, calcite, cuprite, calamine, azurite, smithsonite, malachite, chromite, arsenic trioxide, orpiment, kaolinite, montmorillonite, arsenopyrite, pyrolusite, nepheline, andesite, etc. [11, 12, 13]. The sulfide ore deposition also rises as it is connected with the mineralization of the gold and hydrous iron oxide ($\text{Fe}_x\text{O}_y \cdot n\text{H}_2\text{O}$) rocks [14]. In this investigation, to assess factors affecting the level of trace metals in groundwater and identification their possible sources and mineralization processes, multivariate statistical techniques, including Pearson's correlation coefficient, principal and robust components analysis, and dendrogram cluster analysis are used.

2. Methods and Materials

2.1 Study area

The investigated area is part of Kushtia District, which is positioned at $23^{\circ}41'$ and $24^{\circ}11'$ north latitudes and $89^{\circ}22'$ east longitudes. The total study zone is 1620.5 sq km and is bounded by the Padma River (Ganges River) and the other three branched rivers making a big deltaic flood plain (Fig. 1). The populace of that area is about 2.5 million and the majority of the inhabitant involved in farming activities [15]. The soil characteristics in this zone are very fertile with nutrient-rich sediments as the area lies in the floodplain of the Ganges River. The top-soil characterizes plain geomorphology which appears to level landscape with a raise of around 9 m above sea level, but in some parts, it contains shallow depression and slightly higher edges [16]. This area is enclosed by subtropic humid weather with warm and rainy in the monsoon and marked dry periods in the winter season. The maximum temperature is mostly detected in May-June and the lowest in December-January. The study area received a total rainfall of 1165 mm per year. About 95% of groundwater is utilized for irrigation events and the remaining for ingesting as drinking water.

The primary aquifer of the study area contains unconsolidated

alluvial sediment which is spread over the top surface by the porous sand, silt, and clayey soil. The geology of the study area comprises young alluvial deposits, deltaic silt deposits, stream, and flood-plain deposits, calcareous sandstone, and conglomerate.

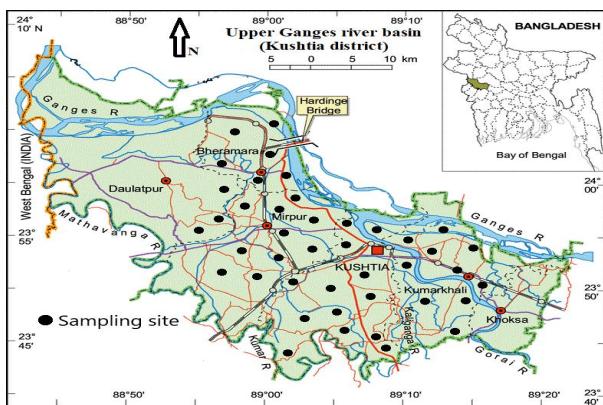


Figure 1: Study area and sampling sites.

According to sub-surface hydrogeological information, it seems that the most important good aquifer in this zone occurs between 20 to 150 m depth from the surface layer. The deepness of the local aquifer varies because of the influences of basement rock thickness and the crosswise extent of the aquifer. The course of aquifer water flow in the study zone is usually from east-north to west-south.

2.2 Sampling strategy

The selection of sampling locations, collection, transportation, type of parameters, and analytical methods of the groundwater samples are the important factors for the present study's rational sampling and analytical strategy. The sampling area is rural and peri-urban. The sampling strategy is based on the identification of trace metal sources and justifies the water quality for drinking purposes. The nonpoint metal pollution sources (diffused sources) are the most common compared to the point sources (single source) in sampling areas. Nonpoint source pollution is the leading cause of water pollution in Bangladesh because it is challenging as there is no single identifiable source. In the study area, runoff and flooding that has accumulated contaminants can sweep sediment, surface pollutants, nutrients, pesticides, fertilizer, inorganic pollutants, livestock wastes, human excreta, etc., into water bodies. Most of the

portions of these matters are leached into the subsoil and finally reach the ground aquifers. With other causes, the overall water quality of the study area is deviated by the above external sources. Table 1 represents the sampling descriptions and probable pollution sources of the group of sampling stations.

For physical and chemical analysis of groundwater, three sampling periods viz. April-May (Pre-monsoon), July-August (Monsoon), and October-November (Post-monsoon) were selected. Groundwater was collected from the shallow aquifer of both residential and agricultural areas, which was up to approximately 100 m below the surface, less than 100 years old, and continuous recharge by rainwater and river streams. Samples were collected randomly from the selected hand pump well to cover the topographical extension of the study area and the key geologic sceneries. It was collected from two different zones viz., residential and agricultural areas, and the depth of sampling wells ranged from 22 to 110 m.

2.3. Analytical procedure

A 40 groundwater sampling sites in the middle-western area of Bangladesh (Fig. 1) were selected for this study throughout the pre-monsoon (PRM), monsoon (MON), and post-monsoon (POM) periods in 2019-2020 and 2020-2021. Samples were collected from each administrative sub-zone of the 4 Upazilas of Kushtia district. Water samples were taken out in washed high-density polyethylene (HDP) plastic flasks as stated by the usual procedure [17]. After 3-5 min. of pumping, the water samples were collected to avoid any debris. For trace metal investigation, the samples were well-preserved with AR grade HNO₃ and kept at 4°C for further analysis. The study measured a total of 11 trace metal parameters such as iron (Fe), manganese (Mn), nickel (Ni), chromium (Cr), cadmium (Cd), lead (Pb), cobalt (Co), arsenic (As), boron (B), copper (Cu), and zinc (Zn) with some associated physicochemical water parameters such as pH, total hardness (TH), electrical conductivity (EC), and total dissolved solids (TDS). Instantly pH, EC, and TDS were measured in the sampling field by the portable multimeter, and TH was estimated by the general titrimetric method. Trace elements were analyzed by the well-

Table 1: Sampling descriptions and probable metal and other pollution sources of the sampling stations.

Sampling field	Sample ID	Point sources	Nonpoint sources
Residential area	1, 2, 3, 4, 5, 6, 7, 8, 11, 18, 19, 23, 24, 26, 32, 35, 36, 37, 38, and 39.	- On-site septic systems, leaking septic tanks - Landfill - Livestock wastes	- Atmospheric deposition and hydrologic modification - Microbes and nutrients coming from pet washes, livestock, and damaged septic systems - Drainage, seepage - Flooding or overflow - land runoff - Soil erosion and sediment runoff
• Shallow well (22-65 m)			
• Relatively high land			
• Mostly in river valley area			
Agricultural area	9, 10, 12, 13, 14, 15, 16, 17, 20, 21, 22, 25, 27, 28, 29, 30, 31, 33, 34, and 40.	- Livestock wastes - Landfill	- Land runoff, flooding, waterlogging - Chemical pesticides and fertilizers that infiltrate the soil and make their way into an aquifer - Soil washed and erosion from heavy farming - Contaminants in rainwater - Atmospheric deposition and hydromodification - Organic wastes from farmlands
• Shallow/Semi-deep well (40-110 m)			
• Mostly in deltaic basin area			
• Relatively low land			

accepted method through Atomic Absorption Spectrophotometer (AAS: Perkin-Elmer Model 3110). Quality control of investigation was kept in all metal analyses as stated by distinct instruction manuals and method accuracy was more than 95% in confidence interval (CI) with the correlation coefficient, $r = \sim 1$ of individual calibration curves.

2.4. Statistical and geostatistical analysis

Several statistical techniques such as Pearson's correlation matrix, robust and principal component analysis (PCA), Hierarchical cluster analysis (HCA), bivariate plot, etc. were used to evaluate the source of solutes in the groundwater solution. All the statistical computations were accomplished using IBM-Excel Worksheet, IBM-SPSS v-26 programs, and geostat v-2. Data screening exhibited that the data are normally skewed. Consequently, the data sets were log-transformed and resembled more closely to normally distributed data. At that time, all variables were standardized by computing their standard scores (z-scores). The PCA and the factor analysis are different types of

analysis. While these analyses are ordination statistical processes that are the primary methods to recognize the geochemical weathering that designates the aquifer mineralization. These are discriminating the factors like manmade and geogenic processes that impact groundwater chemistry [18, 19, 20]. The factors are self-determining, and varimax rotation was exploited to recognize factors encompassing the highest (normally distributed) variability; the variability is implied as an eigenvalue [21, 22]. Factors are considered with eigenvalues greater than one, thus limiting the number of factors used in the PC analysis. Cluster analysis (CA) is a valuable tool to organize samples into classified collections. The Q- and R- modes HCAs of the water samples were presented to the outline groups of samples with the content of geochemical parameters and provided helpful statistics from the component analysis.

4. Results and Discussion

4.1. Trace metal distribution in groundwater samples

Trace metals are a widespread limit of contaminations (mg/L) and steady a human health concern owing to their toxicity

dimensions even in low concentrations and can show an opposite impact on living existences with whole ecosystems and a tendency to biomagnification and bioaccumulate in lipids, fat, or tissues of organisms over time being [23, 24]. The metals such as Cr, Co, Cd, Pb, Hg, and As have no essential function in human physiology. Furthermore, long-term exposure may cause acute disruptions in the normal operations of the human organ systems where those metals are accumulated [6]. Several trace metals, like Fe, Mn, Cu, and Zn, play as micronutrients and are required in the human body in limited quantities for metabolic actions, but, at a higher level can cause opposite health effects [4]. The main manmade sources of trace elements in groundwater are natural substances percolated into the soil system or rocks, the residue of agrochemicals, precise release from the industrial run-off and sewage treatment plant, and uncontrolled escape from landfill spots and chemical accidents or disasters [25]. The groundwater is contaminated with excessive trace metals, particularly arsenic, and has become an alarming situation in Bangladesh. A total of 11 trace metals were investigated during three sampling seasons and the obtained results are stated in Table 2. On the other hand, Fig. 2 shows the average concentration of these metals in three sampling periods. The result showed that the metal concentrations were usually obtained in the order of the PRM<MON<POM sampling periods. The standard deviation ($\pm SD$) of all data is found to be very poor, and a low standard deviation means data are clustered around the mean concentrations. Besides, the skewness values showed that the data is normally distributed [26].

The study results revealed that an average of 100% of water samples carried an excess level of Fe and Mn in all sampling periods. The mean Fe and Mn concentrations of the three sampling periods were 8.435 and 2.848 mg/L, which are much greater than the permissible limit (Table 3). So, Fe and Mn contaminated water could seriously threaten public health and irrigation in the study area. On the other

hand, same Table 3 illustrated that analyzed results showed that around 50% of samples contained overloaded As (0.009 mg/L), Co (0.045 mg/L), Ni (0.320 mg/L), Cr (0.047 mg/L), and Cd (0.008 mg/L) than threshold limits. But the average concentration of Zn and Cu in groundwater in the study area was found 4.027 and 1.693 mg/L, of which 70.9 and 34.1% of samples cross the tolerance level, respectively (Table 3).

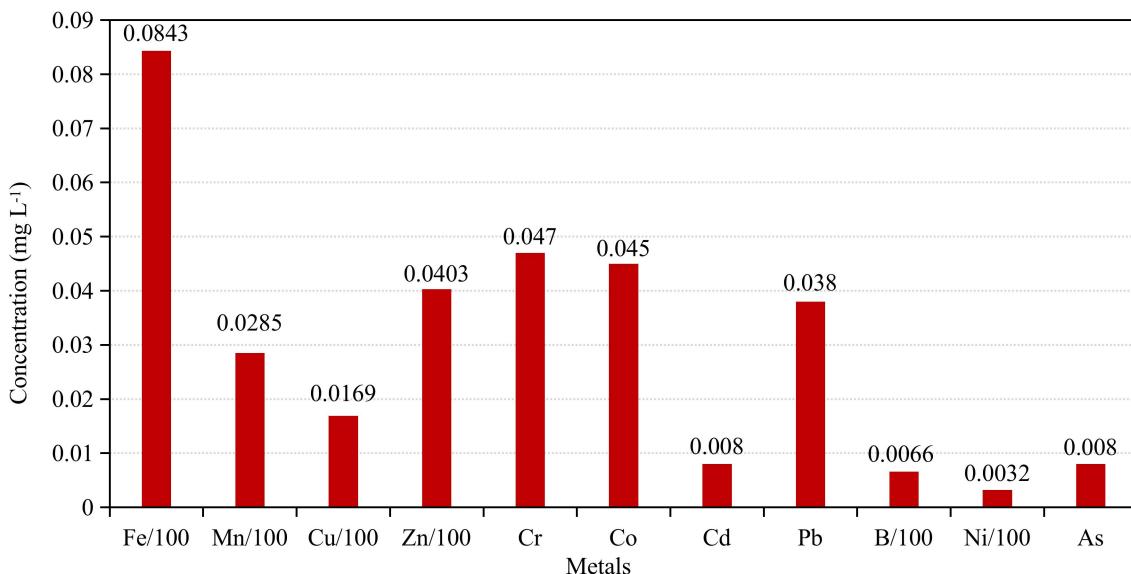
4.2. Source rock of metals in an aquifer

The bioavailability of metals in groundwater depends on various physical, lithological, chemical, and bacteriological factors in sedimentary aquifers. The availability of metals at the sediment level is closely tied to the quantity of sediment-laden trace metal dissolved in neighboring pore water. In oxic sediments, many metals may be adsorbed on the surface of clay particles; and Fe, Mn, and Ni oxides are coated on clay or organic particles [27]. As the level of O₂ in sediment reduces, typically due to bacterial degradation, the metal-oxide coatings begin to dissolve, liberating adsorbed metals. In anoxic sediments, several metals react with sulfides produced by microbial actions to form unsolvable metal sulfides [28]. Metals may be unconfined from sorbet into sediment pore water in ionic forms during changes in pH and oxidation-reduction potential (Eh). For example, in oxic-sediment, Cu and Ni are adsorbed on the surface of organic matter (OM) with Fe/MnO and only Fe/MnO respectively. In anoxic sediment, these two metals (Cu and Ni) are adsorbed on the surface of sulfide rocks of Cu and Fe (Cu₂S, CuS, FeCuS, etc.) and OM/NiS respectively [29]. Also, adsorbed metals may release into pore water by microbial degradation of organic matter. Certain microbes can methylate some metals, such as mercury, arsenic, and lead, into organic types that are more available than inorganic forms [30]. Except for the geogenic mechanism of metal dissolution in groundwater, metal concentrations can increase by anthropogenic causes. Based on the average metal concentration of different sampling seasons, some statistical methods are used here to find out the corresponding source of each trace metal.

4.3. Sources and dissolution of trace metals

Table 2: Statistical analysis of trace metals in the groundwater samples of the Pre-monsoon (PRM), Monsoon (MON), and Post-monsoon (POM) sampling seasons.

Pre-monsoon (PRM)											
	B	Fe	Mn	Cr	Pb	Co	Ni	Cd	As	Cu	Zn
Min.	0	1.14	0.66	0	0	0	0	0	0	0.03	0.79
Max.	1.25	12.62	4.84	0.088	0.096	0.088	1.095	0.051	0.019	3.11	6.63
Mean	0.631	7.997	2.725	0.043	0.035	0.043	0.304	0.008	0.009	1.592	3.940
SD (\pm)	0.394	3.156	1.227	0.024	0.034	0.023	0.351	0.011	0.006	0.831	1.618
Skewness	-0.416	-0.438	0.115	-0.397	0.482	-0.476	1.132	2.131	0.077	-0.108	-0.273
Monsoon (MON)											
Min.	0	1.87	0.46	0	0	0	0	0	0	0.14	0.79
Max.	1.235	13.73	5.66	0.088	0.092	0.097	1.185	0.042	0.019	3.32	6.72
Mean	0.661	8.521	2.872	0.048	0.038	0.045	0.324	0.008	0.009	1.732	4.047
SD (\pm)	0.395	3.273	1.373	0.023	0.031	0.025	0.358	0.011	0.007	0.865	1.584
Skewness	-0.374	-0.308	0.143	-0.665	0.328	-0.362	1.168	1.979	-0.089	-0.091	-0.365
Post-monsoon (POM)											
Min.	0	1.985	0.540	0	0	0	0	0	0	0.090	0.670
Max.	1.450	14.440	5.605	0.095	0.099	0.093	1.190	0.045	0.022	3.370	6.770
Mean	0.692	8.783	2.944	0.051	0.040	0.047	0.332	0.008	0.009	1.755	4.085
SD (\pm)	0.419	3.3503	1.360	0.023	0.031	0.024	0.355	0.012	0.007	0.880	1.607
Skewness	-0.170	-0.443	0.09	-0.550	0.269	-0.570	1.145	2.008	0	-0.110	-0.410

**Fig. 2.** Average metal concentrations (mg/L) of three sampling seasons. The concentrations of Fe, Mn, Cu, Zn, B, and Ni are denoted by 100×mg/L.

The presence of trace metals in the groundwater can be due to geochemical, geogenic, or human activities. In natural or geogenic activities, the dissolution of minerals or rocks that contain these metals, soil-water interaction, an explosion from volcanos, deposition of atmospheric salts, and geological occurring processes of dissolution can be comprised [31]. From these happenings, trace metals enter dissimilar units of the environment. Trace metals can be

found in the forms of sulfates, sulfides, oxides, hydroxides, phosphates, and silicates or mixed types [32]. Although the rapid increase in industrialization and urbanization is an example of manmade sources through which water gets polluted by trace metals. The local geology, agrochemicals leaching, overexploitation of groundwater, etc., were the major causes of toxic metals in drinking water [33, 34, 35]. The metal concentration in groundwater sometime depends

Table 3: Statistics of trace metal compositions in groundwater during the PRM, MON, and POM with the respective standard values.

Metal	Pre-monsoon, PRM (n = 40)		Monsoon, (MON) (n=40)		Post-monsoon, POM (n = 40)		Mean value of three seasons	Average %Samples crossed the WHO std.	The MLC value for drinking water		
	Mean value	%Samples exceed the WHO std.	Mean value	%Samples exceed the WHO std.	Mean value	%Samples exceed the WHO std.			WHO [45]	USEPA [17]	DPHE ^a [16]
B	0.631	0	0.661	0	0.692	0	0.661	0.0	2.4	None	1.0
Fe	7.997	100	8.521	100	8.783	100	8.434	100.0	0.3	0.3	0.3-1.0
Mn	2.725	100	2.872	100	2.944	100	2.847	100.0	0.1	0.05	0.1
Cr	0.043	52.5	0.048	47.5	0.051	37.5	0.047	48.7	0.05	0.1	0.05
Pb	0.035	60	0.038	70	0.040	80	0.038	70.0	0.01	0.015	0.05
Co	0.043	42.5	0.045	47.5	0.047	55	0.045	48.2	0.05	None	None
Ni	0.304	50	0.324	55	0.332	55	0.320	53.2	0.1	0.1	0.1
Cd	0.008	47.5	0.008	50	0.008	47.5	0.008	48.4	0.003	0.005	0.003
Cu	1.592	32.5	1.732	35	1.755	35	1.693	34.1	2.0	1.3	1.0
Zn	3.940	72.5	4.047	72.5	4.085	67.5	4.025	70.9	3.0	5.0	5.0
As	0.009	47.5	0.009	50	0.009	52.5	0.009	50.0	0.01	0.01	0.05

Table 4: pH, EC ($\mu\text{S}/\text{cm}$), TDS (mg/L), and TH (mg/L) values in different 3 sample seasons.

Period	pH range (mean)	EC range (mean)	TDS range (mean)	TH range (mean)
Pre-monsoon (PRM)	6.63-8.04 (7.03)	367.5-1043.5 (670.0)	220.0-667.5 (413.4)	121.5-564.0 (362.8)
Monsoon (MON)	7.06-8.41 (7.43)	560.5-1237.0 (867.5)	369.0-815.9 (558.2)	157.1-594.4 (396.1)
Post-monsoon (POM)	7.14-8.91 (7.80)	663.0-1710.0 (947.1)	449.0-1108 (600.6)	225.7-613.3 (404.8)

Table 5: The average concentration (mg/L) of analyzed trace metals and some selected parameters in the groundwater samples of the three sampling seasons with basic statistics.

	Depth	pH	EC	TDS	TH	B	Fe	Mn	Cr	Pb	Co	Ni	Cd	As	Cu	Zn
Min.	22	7.03	530.3	351.8	216.6	0	1.71	0.55	0	0	0	0	0	0	0.09	0.75
Max.	125	8.22	1305.3	866.8	576.5	1.3	13.3	5.25	0.089	0.095	0.09	1.137	0.045	0.019	3.27	6.71
Mean	50	7.43	831.3	528.2	387.9	0.66	8.43	2.85	0.047	0.038	0.045	0.320	0.008	0.008	1.69	4.03
SD(\pm)	31.9	0.23	172.8	110.4	84.04	0.40	3.23	1.30	0.023	0.032	0.024	0.353	0.011	0.006	0.85	1.60
Skewness	0.96	0.76	0.82	1.05	0.33	-0.34	-0.41	0.12	-0.51	0.38	-0.45	1.15	2.04	-0.08	-0.12	-0.35

on the several physicochemical parameters of water samples such as pH, EC, TDS, and TH. Besides, water depth is another factor that regulates the mineralization of aquifer and solute concentration. The values pH, EC, TDS, and TH in different 3 sample seasons viz. pre-monsoon (PRM), monsoon (MON), and post-monsoon (POM) periods are presented in Table 4. Also, the average concentration of trace metals and some relevant physicochemical parameters, including water depth, pH, EC, TDS, and total hardness (TH)

of the three seasons are stated in Table 5. The dataset indicated that the maximum parameter values in the MON and POM seasons were higher than in the PRM season, which may cause percolation leaching during the rainy season in the study areas (Tables 4 and 5). In this section, the identification of metal pollution sources is performed through the source rocks in the aquifer and using multivariate statistical techniques.

4.4. Multivariate statistical analysis

Various statistical and computer programs were used to source

and dissolve trace metals in local aquifer groundwater. In this connection, multivariate analysis such as R and Q mode Cluster analysis, Pearson's correlation matrix, and principal component analysis, numerous biplots, diagrams, and programs are used for this purpose and are stated below:

(i) Pearson's correlation matrix

Pearson's correlation matrix was utilized to realize possible correlations between these trace metals to determine whether either the presence of specific trace metals in a sample facilitates the occurrence of other metals, or if they coexist owing to anthropogenic or geogenic activity in the study areas. Table 6 summarizes the inter-parameters correlation coefficient in groundwater. The inter-metal and metal with other relevant parameter relationships deliver useful information on metal sources and flow paths [36]. The correlation analysis displayed strong and significant positive correlations ($r>0.7$) in some trace element pairs, such as Fe with Mn, Co, Pb, and Zn; Mn with Fe, Pb, Co, Cu, and Zn; Pb with Fe, Mn, Cd, Cu, and Zn; Co with Fe and Mn; Cu with Fe, Mn, Pb, and Zn; and Zn with Fe, Mn, Pb, and Cu. Other metals are not strongly correlated with others. It was seen that maximum metals are strongly correlated with Fe and Mn. In the case of other physical parameters, all most all-metal concentrations are negatively correlated with water depth, i.e., the concentration of trace metals decreases with the aquifer depth. The total hardness (TH) does not correlate with any trace metal, such a result indicated that Fe or any other trace metal is not responsible for water hardness. The EC and TDS are strongly positively associated with the Fe, Mn, Cd, Cu, and Zn concentrations. So, these metal concentrations depend on salinity and the mineralization rate of the groundwater [37]. The pH is slightly negatively correlated with all metals, which indicates that in lower pH (acidic medium) the metal concentrations are increased by a higher dissolution rate [38]. The dissolution of metal ions in aquifers mostly depends on water pH. At a low pH value, the trace metals tend to be more dissolution due to more solubility in an acidic water solution [39, 40]. Another study by these authors [41] showed that the pH values were acidic

in the maximum water samples in the PRM season, indicating fewer metal ions present in higher concentrations compared to the POM season (Tables 2 and 4). But dataset showed that the average metal concentration of the PRM was lower than the MON and POM sampling seasons indicating other geogenic causes are responsible rather than the water pH (Table 2).

Based on the lithology of the study zone, the coarse sandy alluvial geologic form, and the deltaic floodplain land are thought to be porous and penetrable enough to allow the passage of the trace metal-laden water into the aquifer basement [42]. The correlation matrix showed that most of the trace elements have substantial relations with each other, indicating the similarity of sources [43]. It illustrated that Fe and Mn significantly correlate with all the trace metals and that the presence of extra Fe and Mn in the samples can be attributed to geogenic and manmade processes. The geogenic processes that could release Fe and Mn in groundwater include the redox condition of the aquifer systems and the rusting properties of water supply pipes [44].

Among the eleven (11) analyzed heavy metals, the Fe, Mn, Ni, Pb, Cu, and Zn levels of more than 50% of samples exceeded the WHO standard permissible level for drinking purposes (Table 3) and they are strongly correlated with each other. The Cd and Pb may be coming from the same contamination source due to their strong positive correlation. Cd occurs naturally with Pb in the same sulfide ores [45]. B, Cd, Cr, As, and Co have weak correlations with each other, and the dissolution processes occurred from different sources. The absence of correlation between the metal elements proposes that the contents of these elements are not measured by a unique factor, but rather by an amalgamation of hydro-geological support phases [46]. Loading association and cluster characteristics of the metal concentration are discussed in the PCA and cluster analysis

(ii) Principal component analysis (PCA)

Principle component analysis (PCA) and robust PCA were utilized to compare the compositional and spatial outlines between the investigated water samples and the recognized latent factors and find the probable sources of the trace .

Table 6: Person's correlation matrix for trace metals and physicochemical parameters (from SPSS software).

	Depth	pH	EC	TDS	TH	B	Fe	Mn	Cr	Pb	Co	Ni	Cd	As	Cu	Zn
Depth	1.00															
pH	-0.22	1.00														
EC	-0.08	-0.40	1.00													
TDS	-0.10	-0.44	0.98	1.00												
TH	0.11	-0.35	0.70	0.70	1.00											
B	0.24	-0.15	0.12	0.14	0.18	1.00										
Fe	-0.05	-0.31	0.57	0.53	0.29	0.17	1.00									
Mn	-0.03	-0.25	0.60	0.55	0.26	0.15	0.89	1.00								
Cr	0.13	-0.24	0.34	0.35	0.29	0.31	0.40	0.32	1.00							
Pb	-0.11	-0.25	0.46	0.43	0.06	0.10	0.64	0.70	0.39	1.00						
Co	-0.07	-0.02	0.25	0.19	0.08	0.26	0.64	0.75	0.20	0.37	1.00					
Ni	-0.11	-0.23	0.41	0.40	0.24	0.25	0.35	0.47	0.05	0.46	0.18	1.00				
Cd	-0.05	-0.35	0.50	0.51	0.32	0.32	0.41	0.46	0.35	0.50	0.25	0.24	1.00			
As	0.17	-0.14	0.42	0.38	0.56	0.31	0.41	0.41	0.29	0.25	0.23	0.34	0.26	1.00		
Cu	0.02	-0.30	0.58	0.56	0.16	0.04	0.72	0.72	0.28	0.78	0.34	0.44	0.38	0.29	1.00	
Zn	0.17	-0.28	0.66	0.63	0.37	0.17	0.77	0.75	0.47	0.57	0.41	0.29	0.44	0.33	0.76	1

Table 7: Principal component analysis of detected water parameters (sorted by size). The table got from SPSS software.

	PC1	PC2	PC3	PC4
Mn	0.869	-0.370	0.081	0.093
Fe	0.853	-0.288	0.159	-0.054
Zn	0.838	-0.101	0.081	-0.252
EC	0.832	0.283	-0.365	0.020
TDS	0.807	0.319	-0.390	-0.007
Cu	0.792	-0.320	-0.112	-0.262
Pb	0.721	-0.410	-0.051	-0.146
Cd	0.612	0.132	0.008	0.026
As	0.539	0.337	0.208	0.388
Ni	0.529	-0.053	-0.181	0.342
Co	0.527	-0.457	0.333	0.340
Cr	0.512	0.169	0.338	-0.201
pH	-0.454	-0.372	0.037	0.425
TH	0.540	0.701	-0.166	0.186
Depth	-0.041	0.343	0.647	-0.429
B	0.297	0.283	0.610	0.355
Variance (%)	42.174	11.501	9.123	6.969
Cumulative (%)	42.174	53.675	62.798	69.767
Eigenvalue	6.748	1.840	1.460	1.115

Note: Bold values indicate strong loading with component number.

elements in the groundwater PCA is extensively applied to classify anthropogenic contributions and geogenic pollution sources. Factor analysis (Table 7 and Fig. 3) gives four PCs of R-mode factors that have eigenvalues greater than one. These four factors have a percentage variance of 42.174, 11.501, 9.123, and 6.969% with Eigenvalue >1. FC1 shows Mn(0.869), Fe(0.853), Zn(0.838), Cu(0.792), and Pb(0.721) have high positive factor values (>0.7) indicating those metals are strongly associated with each other and dissolve from a similar type of source that is possibly exogenous

discharge [47]. Though the origin of Fe and Mn is partially attributed to geogenic processes in the correlation analysis, this class is made up of parameters unusual to manmade sources rather than mineral dissolution. Based on the geological situation of the study area, there have been reports of mineral or soil deposits rich in trace metals that could release these potentially toxic elements into the aquifer systems [48, 49]. It was assumed that the above-mentioned highly associated metals are dissolved with water in aquifer sediment levels from the mixed rock such as (FeNi)O(OH), (CoFe)AsS, ZnCrO₄,

CuFeS_2 , $(\text{FeZn})_6\text{Sb}_2\text{S}_9$, etc. [50]. The Cd, As, Ni, Co, and Cr have moderate factor values in PC1, which indicates they are moderately associated with each other. In PC2 and PC3, the factor value of the maximum metals is near zero or negative. Only boron (B) has a significant factor value in PC3 (0.610). The negative factor value of water depth and pH indicates the negatively associated with all the metal concentrations, which were supported by Pearson's correlation matrix.

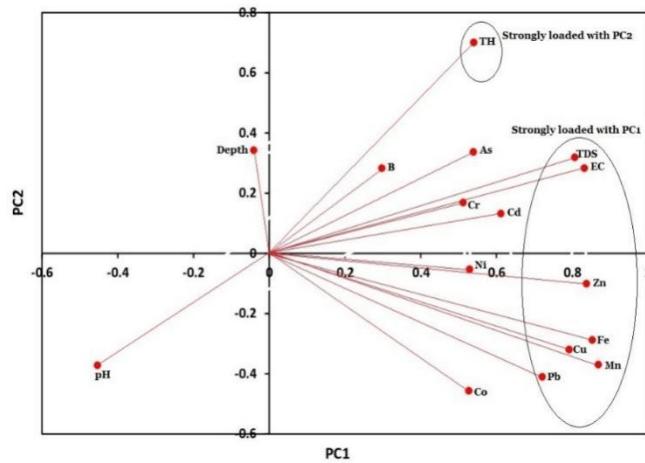


Figure 3: Robust PC of metals and other selected water parameters in the water samples (Fig. plotted with the help of Microsoft excel and Aqua-Chem software).

The values with positive and negative in PCA explained that the water samples were affected or unaffected by the occurrence of extracted loads on an exact component. The moderately strong association or correlation among EC, TDS, and TH indicates the presence of a huge ionic form of the metal component which is gathered by aquifer rock-water interaction and manmade sources, like agrochemicals from agricultural run-off [51]. The results of the PCA were observed by statistical cluster analysis and TDS vs metals plot, which is discussed in the next sub-sections.

(iii) Cluster analysis

The hierarchical cluster analysis method gives 3 (three) major cluster groups based on an R- and Q-mode dendrogram with Ward's method for the trace metals with some physical parameters in the study area (Fig. 4). Parameters that fit in a similar cluster are probable to have been leached from the same mineral/rock source [52]. Like PCA, cluster analysis places parameters into groups based on illustrious same

features and relations with each other. In an agglomerative schedule cluster study, the most alike variables are mentioned in one single cluster group and related to a closely associated cluster(s) and further from clusters with fewer comparative, all of which are related to form one big cluster group [21].

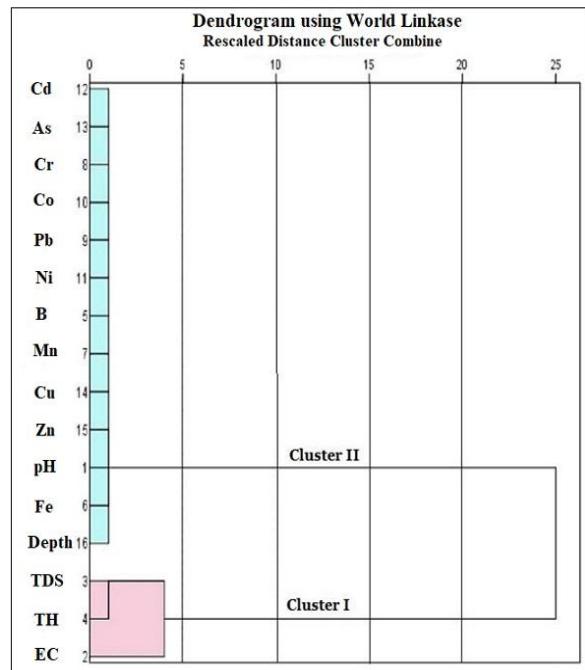


Figure 4: Hierarchical cluster analysis (HCA) of 40 sampling sites for the average value (mg/L) of trace metals and other selected parameters in three sampling seasons (PRM, MON, and POM). Dendrogram of Q-mode HCA presenting associations between samples from different portions of hydrologic systems. Figure constructed from SPSS software.

Cluster 1 is formed by only three variables TDS, EC, and TH. While all measured metals and well depth constructed cluster II. Like the findings from PC and correlation analysis, all-metal concentrations depend on pH and water depth (Cluster II). Low pH and shallow depth are responsible for higher concentrations of metals (mainly Fe, Mn, Pb, Cu, and Zn). The first cluster illustrates an analogous association between total hardness (TH) and total dissolved solids (TDS) and suggests the domination of groundwater by precipitation and associated contact with air CO_2 [36]. Figure 4 shows that EC, TDS, and TH are not in the same cluster as metals indicating the metal loadings are not dependent on other general cations and anions (e.g., Na, K, Ca, Mg, HCO_3^- , SO_4^{2-} , NO_3^- etc.) of groundwater.

It may be assumed that those metals are dissolve from rock

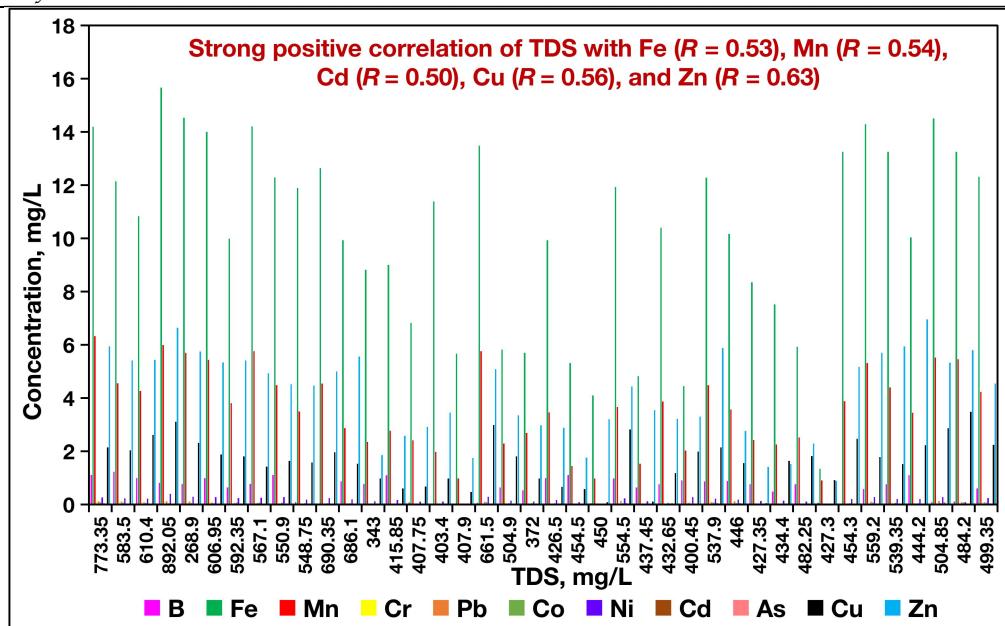


Figure 5: Bivariate diagram of TDS vs trace metals.

water independently and some portion comes from anthropogenic sources such as agrochemicals leaching.

4.5 Mineralization process

The rock/mineral-water interaction and drive of solutes in aquifers systems are the important factors controlling the groundwater mineralization courses. A diagram (Fig. 5) of measured trace metals associated with total dissolved solids (TDS) shows that the geochemical facies took part in the groundwater mineralization [53]. This Fig. 5 establishes that the analyzed trace metal ions, Fe ($r=0.53$), Mn ($r=0.55$), Cd ($r=0.51$), Cu ($r=0.56$), and Zn ($r=0.63$) of groundwater are strongly positively correlated with TDS. This result represents the leading trace metals that donate to the groundwater through metal-laden sediment in the aquifer. Water chemistry indicated that Fe, Mn, Pb, Cu, and Zn are the most abundant trace metals in the study zone's groundwater samples. These results endorse the constant addition of these metal ions along the aquifer water flow path also suggestively contributes to groundwater mineralization. The Fe and Mn concentrations crossed the guideline value in the 100% samples (Table 2). These two ions may be invented from similar sources. Thus, dissolution or weathering is the geogenic geochemical process controlled by the salt load along the groundwater

flow path [54].

The four statistical methods mentioned above, viz, correlation matrix, PCA, cluster analysis, and bivariate regression correlation, are used to identify trace metal pollution sources. Pearson's matrix revealed that all the metals are moderately or weakly correlated in the acidic medium of water samples. The study area is an agricultural zone and deeply irrigated using shallow groundwater. No, an industrial plant is situated in the study area or neighboring area. So, among the anthropogenic source, only agrochemical leaching is the cause of metal sources in groundwater. Besides, several studies assumed that the trace metal loading in aquifers occurred by local geogenic activities which are positively influenced by heavy water mining [55, 56, 57]. Therefore, except for some anthropogenic sources, the geogenic source is the key factor in increasing the trace metal concentration in aquifers and was supported by above mentioned statistical analysis.

5. Recommendations

The chemical features of groundwater in the study area were dependent on the water-rocks interaction, carbonate-based mineral dissolution, and most neighboring river morphology. The study findings may be used for sustainable water

resources management on a regional scale and may be put in another area with similar topography. Accordingly, it is expected that the results of this investigation would deliver useful understandings for future groundwater monitoring and management of the study area. Also, the findings of this investigation deliver a guideline for agricultural activists, policymakers, public health departments, and water managers of a shallow aquifer. In addition, the study suggests that the groundwater should be drunk after proper treatment for removing the trace heavy metal contaminant from the potable water. Besides, deep wells should be drilled near river valleys and avoid thick clay-layer areas.

This study measured higher levels of some trace metals in samples that make the water very harmful for every purpose. Elevated concentrations of iron, manganese, and lead seriously impacted the drinking water quality. Further study would be considered using modern and sophisticated investigative approaches with extensive parameters over a wide area. The study found that the lack of public awareness of water quality, inadequate modern water treatment practices, absence of water quality monitoring, as well reluctance of law implementation are the main challenges to safe water supply in the study area. Moreover, public awareness building and publicity programs towards groundwater contamination are imperative to ensure safe water for all. Further, advanced research and survey-based works in the areas needed to be enhanced to explore the safe water status and water-related problems.

According to the research findings, some recommendations to manage the sustainability and purposes of groundwater resources are proposed as follows:

- The water from the deeper aquifer is more suitable than the shallow aquifer for both irrigation and domestic purposes and should be used in deeper water instead of shallow water.
- A field study confirmed that the hardness and trace metal level of single-owner water-well is generally higher than those of community users. So, this study suggests the local government should ensure a

community-based water supply system instead of a single-owner system.

- The groundwater should be drunk after 24 hrs. of extraction.
- The low-cost and indigenous materials made for filtration with solar disinfection processes could be used to reduce the temporary hardness, some toxic metals, and microbes.
- Obsessive extraction of groundwater needs to control.
- It should be frequently monitored the chemical and trace metal characteristics of groundwater.

6. Conclusion

Trace elements in groundwater can have both natural and manmade sources due to industry, industrial effluents, and agrochemicals. In this study, the concentration of trace metals was determined from 40 groundwater samples within three seasons in the study area of the middle west part of Bangladesh. The analysis result showed that the maximum metal concentration exceeds the guideline values of the respective component. Three metals ion, viz. Fe, Mn, and Pb crossed the limit of WHO standards in the maximum of the water samples representing severe human health hazards. The levels of Fe (8.43 mg/L) and Mn (2.85 mg/L) were found 28 fold greater than the standard value of 0.3 and 0.1 mg/L, respectively for drinking purposes. Another finding is the trace metal concentration in MON and POM seasons are found to be somewhat higher than PRM. The investigation results exposed that mainly the natural processes and little manmade activities impacted the trace metal loads in the groundwater system of the area. To assess factors affecting the level of trace metals in groundwater samples and determine their probable sources and mobility, multivariate statistical techniques, including Pearson's correlation coefficient, principal components analysis, and dendrogram cluster analysis are utilized. Results revealed that sources of trace elements were identified to be geogenic and slightly from human actions, i.e., application of agrochemicals, excess water mining, and industrial wastewater. In addition, it was found that the level of trace elements in samples

somewhat depends on water pH and salinity. Groundwater in this area poses a huge threat due to high levels of trace metals, and without any purification, it is necessary to avoid this water for drinking.

Authors Contribution

MSI and MGM has the main idea of the manuscript and wrote the the manuscript. MT revised the manuscript and provide suggestions.

Conflicts of Interest

The authors reported no potential conflict of interest.

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Data Availability statement

The data presented in this study are available on request from the corresponding author.

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